Optical Analysis of Coated and Uncoated Soot Particles: Data for Global Climate Change Models

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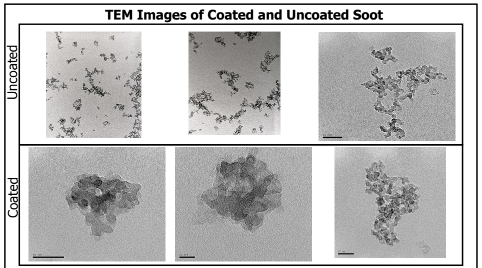
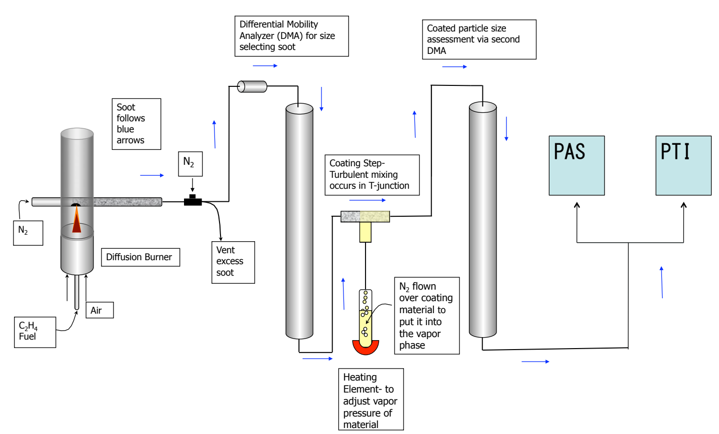
Atmospheric aerosols play a fundamental role in Earth's atmospheric chemistry and climate. Soot is an absorbing aerosol, though the magnitude of that absorption has largely been determined by measuring the optical properties of uncoated soot. It has been proposed that coated soot might absorb radiation more efficiently than uncoated soot, thus warming the climate more than previously suspected. For this study, soot is generated in a well-controlled Santoro-Style diffusion flame burner with ethylene as the fuel, and has been successfully coated with dibutyl phthalate (DBP), a non-absorbing material. DBP has a refractive index of 1.490 (real part), which is similar to the refractive index of sulfuric acid (n=1.426) at 589 nm. DBP is substituted for the commonly found sulfate coated particles for several reasons including safety and instrument integrity. By changing the temperature of the DBP, the vapor pressure of the DBP is changed and consequently, the coating thickness can be changed. Extensive work has taken place in controlling the coating process as well as in the generation and delivery of the aerosols for analysis. The aerosols are classified with a differential mobility analyzer (DMA). From the DMA the aerosols are sent to a condensation particle counter (CPC) for size distributions or are analyzed. Analysis will be performed in a collaborative effort using both a Photoacoustic Spectrometer (PAS) and a Photothermal Interferometer (PTI) system. Both techniques measure absorption directly and this is a vast improvement over techniques that use extinction and scattering to obtain the absorption.

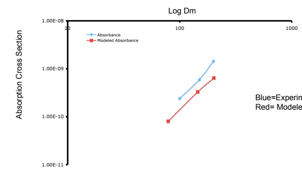
TEM Images of Coated and Uncoated Soot

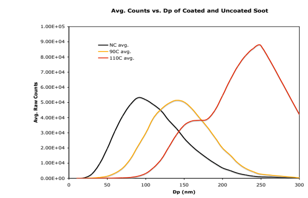


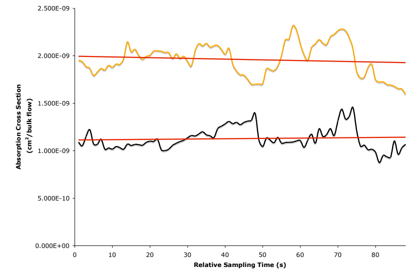
Experimental Design



Modeled and Experimental Results







Results in the graph to the left show peak absorption reached during the sampling period. The black line represents the uncoated soot (ie. pure soot) and the orange line represents the same size soot but coated with a 35 nm coating of the non-absorbing dibutyl phthalate coating. From peak absorbance to peak absorbance an approximate two fold increase in the absorbance. This is solid evidence that non-absorbing coatings on absorbing particles may increase the particle absorption cross section.

Calculations

$$\sigma_{abs} = \text{absorbance/particle}$$
$$\sigma_{extinction} = \text{extinction/particle}$$
$$\sigma_{scattering} = \text{scattering/particle}$$
$$L = \text{path length}$$
$$N = \text{number concentration}$$
$$C = \text{Calibration Factor}$$

$$I_s = \frac{\text{Soot}_{\text{signal}}}{\text{Reference}_{\text{signal}}}$$
$$I_o = \frac{\text{Cal.Std.}_{\text{signal}}}{\text{Reference}_{\text{signal}}}$$
$$\sigma_{extinction} = \frac{-Ln(I_s/I_o)}{L \times N}$$
$$\sigma_{scattering} = \frac{C(\text{Soot}_{\text{scattering}} - \text{Cal.Std.}_{\text{scattering}})}{N}$$
$$\sigma_{extinction} - \sigma_{scattering} = \sigma_{absorbance}$$

Conclusions/Future Work

Preliminary experimental results show an increase of approximately two fold in the absorbance of the coated soot aerosol in comparison to the uncoated soot aerosol. This increase is certainly in the upper ranges of the acceptable bounds but much more work is underway to provide a more accurate result. More experiments are underway and as the measuring technique is improved, so will the value of the absorbance. The results show that a non-absorbing coating on an absorbing aerosol does indeed increase the effective absorption cross section for that aerosol. This effect is a function of the coating's refractive index as well as the absorptive properties of the core aerosol and ultimately the thickness of the coating. Currently, more experiments are being run where the coating thickness is being varied to determine the importance of the coating's thickness and index of refraction with respect to the absorptive properties of the core model.

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Acknowledgements

A very special thanks to Stephanie Winter, Kyle Sullivan, Dominik Cieslack and J. Vanderlei Martins for their help in different areas of this research. We would also like to thank **National Institute for Climate Change Research** for the funding for this project.